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This research sponsored by the SDIO Biomedical and Materials Sciences FEL Program held the following objectives. Provide a facility in which in-house and outside user research in the materials and biological sciences can be carried out in the Far Infrared using the unique properties of the UCSB electrostatic accelerator-driven FEL. Develop and implement new FEL concepts and FIR technology and encourage the transfer and application of this research. Train graduate students, post doctoral researchers and technical personnel in varied aspects of scientific user disciplines, FEL science and FIR technology in a cooperative, interdisciplinary environment. In summary, a free electron laser facility has been developed which is operational from 200 GH<sup>2</sup>, (6.6 cm<sup>-1</sup>), to 4.8 THz, (160 cm<sup>-1</sup>) tunable under computer control and able to deliver kilowatts of millimeter wave and far-infrared power. This facility has a well equipped user lab that has been used to perform ground breaking experiments in scientific areas as diverse as bio-physics. Nine graduate students and post doctoral researchers have been trained in the operation, use and application of these free-electron lasers.

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**Biomedical and Materials Research with the Free Electron Lasers**

**Final Technical Report for the Period**

**1/January 1986 - 31 December 1990**

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## **OVERVIEW**

This research sponsored by the SDIO Biomedical and Materials Sciences FEL Program held the following overall objectives.

- i) Provide a facility in which in-house and outside user research in the materials and biological sciences can be carried out in the Far Infrared using the unique properties of the UCSB electrostatic accelerator-driven FEL.
- ii) Develop and implement new FEL concepts and FIR technology and encourage the transfer and application of this research.
- iii) Train graduate students , post doctoral researchers and technical personnel in varied aspects of scientific user disciplines, FEL science and FIR technology in a cooperative, interdisciplinary environment.

In summary, a free electron laser facility has been developed which is operational from 200 GHz, ( $6.6 \text{ cm}^{-1}$ ), to 4.8 THz, ( $160 \text{ cm}^{-1}$ ), tunable under computer control and able to deliver kilowatts of millimeter wave and far-infrared power. This facility has a well equipped user lab that has been used to perform ground breaking experiments in scientific areas as diverse as bio-physics, bio-medicine, magnetism, semiconductors and laser physics. Nine graduate students and post doctoral researchers have been trained in the operation, use and application of these free-electron lasers.

There were three main objectives of this research program. i) Provide facilities for in-house and outside user research with the far-infrared and millimeter wave FEL's. ii) Develop and implement new FEL concepts and transfer them to applications. iii) Train researchers in the use and relevant science and technology.

### **FACILITIES**

Two reliable Free-electron lasers have been developed that can be used to deliver radiation for a wide variety of scientific experiments; the first covering the spectral range from 313 to 62 microns and the second from 2.5 millimeter to 338 microns. The electron beam is provided by a single 6MV electrostatic accelerator and directed to the appropriate laser by a "switch yard" like beam line. The beam line is computer controlled to allow the operator and user to readily change from one laser to the other and to control the lasing frequency. Under the best conditions the computer can cause the frequency of the laser to scan about a set frequency, covering bandwidths as large as 10% of the set point. Power levels in the kilowatt range are transported out of the FEL vault into the user lab via a computer controlled, evacuated, free space optical transport system.

A variety of instruments have been acquired to facilitate experiments on the FEL. Liquid helium cryostats are available with variable temperature sample space and optical access for far-infrared or optical radiation. Magnetic fields are available to 8 Tesla.

Each of the six optical ports of the FEL far-infrared beam transport system in the user lab is equipped with an optical bench and a supply of FIR optical components. A double monochromator with photon counting detection is available for use with either Raman scattering or detection of the effects of FIR pumping by the FEL.

Since the FEL pulse length is of the order of microseconds, and that of the cavity dump coupler 30 nanoseconds, reasonably fast electronics are available.

Several other lasers can be used in conjunction with the FEL; a Nd:YAG/dye laser, a CO<sub>2</sub> TEA laser with pulsed molecular gas FIR laser and a cw CO<sub>2</sub> pumped FIR ,molecular laser.

A Bomem Fourier transform interferometer with resolution of .002 cm<sup>-1</sup> is available for use in conjunction with FEL experiments. It is equipped with cryogenic and magnetic field capabilities in addition to a liquid He<sup>3</sup> cooled bolometer detector.

The staff of the UCSB Center for Free-electron Laser Studies is balanced to provide continued improvement of the facility and to support ongoing experiments. Two full time staff support operations for users and four staff split their time between development and maintenance and operations support.

## **RESEARCH APPLICATIONS OF THE UCSB FREE-ELECTRON LASER**

### **Bio-physics and bio-medical.**

Experiments in the bio-medical and biophysics arena were carried out in collaboration with researchers at Princeton University and University of California at Irvine. Although different biological systems where investigated they were united by a common theme. Can reaction rates of complex systems be modified by driving the low frequency, far-

infrared, vibrational modes? The experimental answer appears to be yes, but possible models and mechanisms for the coupling are not in hand.

Michael Berns and co-workers from University of California at Irvine demonstrated that 200  $\mu\text{m}$  radiation from the UCSB free-electron laser inhibited DNA synthesis. It is not clear what the mechanism might be but it is speculated that the intense fields from the FEL, resonating with vibrational modes of the DNA molecules, may alter reaction kinetics of these complex systems.

"Inhibition of nucleic acid synthesis in cells exposed to 200 micrometer radiation from the free-electron laser." M. Berns and William Bewley, *Photochemistry and Photobiology*, 46, 165 (1987).

"Free-electron Laser at 200 Micrometers Affects DNA Synthesis in Living Cells", M.W. Berns, W. Bewley, C.-H. Sun, P. Templin, *Proceedings of the National Academy of Sciences of the USA*, 87, 2810 (1990).

Robert Austin and co-workers from Princeton examined reaction rates in three different systems. In the first they probed the geminate recombination of CO photo-detached by white light from sperm myoglobin. Far-infrared radiation from the UCSB FEL enhanced the recombination provided that the specimen temperature was below the glass transition for the glycerol solvent.

"A New Spectroscopy? Spin-Tunneling in Heme Proteins", R. Austin, B. Gerstman, P. Mansky, and M. Roberson, *Biophysical J.* 53, 280a (1988).

"Far-infrared Perturbation of Reaction Rates in Myoglobin at Low Temperatures", R. H. Austin, Mark Roberson and Paul Mansky, *Phys. Rev. Lett.*, 62, 1912 (1989).

"Enhancement of Ligand Binding to Myoglobin by Far-infrared Excitation for a Free-electron Laser", B. Gerstman, M. Roberson and R.H. Austin, *J. Opt. Soc. Am.*, 86, 1050 (1989).

The other biological systems were bacteriorhodopsin and photosynthetic reaction centers. In each case the far-infrared radiation from the FEL stimulated changes in the apparent reaction rates as sensed by an optical probe beam. But at this time models and mechanisms that explained the observed effects are lacking.

"Far Infrared Perturbation of Electron Tunneling in Reaction Centers?", R.H. Austin, Mi K. Hong, C. Moser and J. Plombon, Chemical Physics 158, 473 (1991).

### Magnetics

Saturated magnetic resonance of the impurity mode in associated with Mn in FeF<sub>2</sub> were observed with the FEL. To avoid heating effects a cavity dump coupler was used that enables the user to extract the intra-cavity laser power in  $\approx 40$  nsec. The experiments determine that  $T_1 = T_2 = .29$  nsecs and that energy relaxation proceeds by impurity magnon - phonon decay.

"FEL Far Infrared Study of FeF<sub>2</sub>:Mn", J. Spector, J. Kaminski and V. Jaccarino, Solid State Commun. 63, 1093 (1987).

"Far Infrared Saturation Spectroscopy of the Mn Impurity Mode in Antiferromagnetic FeF<sub>2</sub>", J. Kaminski and V. Jaccarino, to be published Phys. Rev. B.

### Semiconductor electronics.

A far-infrared photo-Hall experiment, performed by researchers at Philips Research Lab, Redhill, Kamerlingh Onnes and Huygens Lab, Leiden and UCSB, demonstrated that the far-infrared radiation excited carriers in a two step process. First they are excited into the conduction band and then further heated by the FEL pulse.

"Far-infrared Photo-Hall Experiments on GaAs:Si", J. Kaminski, J. Spector, T.O. Klaasen, W. Th. Wenckebach and C.T. Foxon, J. Opt. Soc. Am., B6, 1030 (1989).

A collaboration between Battke and Kotthaus from the University of Hamburg and Kaminski and Spector at UCSB demonstrated a novel voltage tunable photovoltaic response in field effect devices with grating gates. Si MOSFET's and GaAs devices demonstrated the effect which resonates at the 2-D plasma frequency. The mechanism responsible for this response is not understood at present. Photon drag or a novel thermo-electric effect are being explored.

"Tunable Photovoltaic Response in Semiconductor Field Effect Devices to Far-infrared Radiation of a Free-electron Laser", E. Batke, J. Kaminski, J. Spector, J. Kotthaus, Appl. Phys. Lett., 53, 131 (1989).

The kinetics of electrons bound to shallow donor impurities in n-GaAs was investigated by saturation spectroscopy using the FEL. The relaxation lifetime and ionization probability of the  $2p^+$  and recombination lifetime of electrons in the  $N=0$  Landau level was determined. Further, the complete saturation behavior of the 1s-continuum transitions was characterized. The research was carried out by Prettl and Weispfenning from the University of Regensburg and Kaminski and Spector at UCSB.

"High Power Non-linear Magneto-photoconductivity in n-GaAs Using the UCSB Free-electron Laser", J. Kaminski, J. Spector, W. Prettl, and W. Weispfenning, Int. J. Infrared and Millimeter Waves, 9, 745 (1988).

"Free-electron Laser Study of the Nonlinear Magneto-photoconductivity in n-GaAs", J. Kaminski, J. Spector, W. Prettl and M. Weispfenning, Appl. Phys. Lett. 52, 233 (1988).

In more heavily doped material the photoconductivity induced by intense FEL radiation exhibits a complicated power dependence that can be successfully modeled by stimulated impurity hopping conductivity. Researchers who collaborated in this experiment came from UCSB, Phillips Research Labs, Redhill, and Kamerlingh Onnes and Huygens Lab, Leiden.



"Impurity Hopping Conductivity in GaAs:Si Induced by FIR Radiation of a FEL", J. Kaminski, J. Spector, W. Th. Wenckebach, 19th International Conference on the Physics of Semiconductors, Warsaw, Poland (1988), p1289.

Intense far-infrared radiation has been shown to broaden and quench free-exciton photoluminescence peaks in AlGaAs/GaAs quantum wells.

"Photoluminescence from  $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  Quantum Wells Quenched by Intense Far-infrared Radiation", S.M. Quinlan, A. Nikroo, M.S. Sherwin, M. Sundaram and A.C. Gossard, to be published, Phys. Rev.

DX centers dominate the transport in Si doped  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  for  $x > 0.22$ . One of the most striking effects attributed to DX centers is persistent photoconductivity.

Remarkably, intense low frequency radiation is able to induce capture of photo-excited electrons by DX centers.

"Far-infrared capture of electrons by DX centers", J.J. Plombon, W.W. Bewley, C.L. Felix, M.S. Sherwin, P. Hopkins, M. Sundaram and A.C. Gossard, to be published Appl. Phys. Lett.

Large, non-resonant second order susceptibility has been measured in heterostructures with the free-electron laser. These heterostructures were comprised of half parabolic wells. The second order susceptibility agreed with model calculations but exhibited a less than quadratic dependence. The latter was due to electrons being ejected from the wells at high power levels.

"Large, Non Resonant, Pump Field Dependent Second Order Nonlinear Susceptibility in GaAs/AlGaAs Heterostructures in the Far Infrared", W.W. Bewley, C.L. Felix, J.J. Plombon, M.S. Sherwin, P. Hopkins, M. Sundaram and A.C. Gossard, to be published Phys. Rev.

### **Outside Users**

A number of outside principal investigators have taken advantage of the UCSB FEL's. By research discipline , they are the following:

#### **Bio-physics**

M. Berns, University of California at Irvine

R. Austin, Princeton University

#### **Semiconductor Physics**

J. Kotthaus, University of Munich

W. Prettl, University of Regensburg

W. Wenckebach, F.O.M. Holland

B.D. McCombe, SUNY Buffalo

#### **FEL Physics**

J. Burghoorn, Holland

### **TRAINING OF GRADUATE STUDENTS AND POST-DOCTORAL FELLOWS**

The following have gained their Ph.D. degrees through the Biomedical and Materials Research with the Free Electron Lasers program.

**Joseph S. Spector, February 1988 - The First Condensed Matter Studies Using the UCSB Free-electron Laser**

**Jann Kaminski, May 1989 - Semiconductor Studies Using the UCSB Free-electron Laser**

Abbas Nikroo, November 1990 - Frequency, Temporal and Spatial Evolution of Large K Magnons in  $\text{MnF}_2$

John Plombon (pending) - Stochastic Ionization of Electrons in GaAs/AlGaAs Heterostructures

Bill Bewley, June 1992 - Second Harmonic Generation in GaAs/AlGaAs Heterostructures in the Far Infrared

#### **PUBLICATIONS PRODUCED IN WHOLE OR IN PART BY THIS PROGRAM**

"FEL Far Infrared Study of  $\text{FeF}_2\text{:Mn}$ ", J. Spector, J. Kaminski and V. Jaccarino, Solid State Commun. 63, 1093 (1987).

"Inhibition of nucleic acid synthesis in cells exposed to 200 micrometer radiation from the free-electron laser." M. Berns and William Bewley, Photochemistry and Photobiology, 46, 165 (1987).

"A New Spectroscopy? Spin-Tunneling in Heme Proteins", R. Austin, B. Gerstman, P. Mansky, and M. Roberson, Biophysical J. 53, 280a (1988).

"Impurity Hopping Conductivity in GaAs:Si Induced by FIR Radiation of a FEL", J. Kaminski, J. Spector, W. Th. Wenckebach, 19th International Conference on the Physics of Semiconductors, Warsaw, Poland (1988).

"High Power Non-linear Magneto-photoconductivity in n-GaAs Using the UCSB Free-electron Laser", J. Kaminski, J. Spector, W. Prettl, and W. Weispfenning, Int. J. Infrared and Millimeter Waves, 9, 745 (1988).

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"Free-electron Laser at 200 Micrometers Affects DNA Synthesis in Living Cells", M.W. Berns, W. Bewley, C.-H. Sun, P. Templin, Proceedings of the National Academy of Sciences of the USA, 87, 2810 (1990).

Ballistic Propagation of Magnons", W. Grill and W. Yen, J. Luminescence, 45, 130 (1990).

"Demonstration and Applications of a FIR Cavity Dump Coupler at the UC Santa Barbara FEL Facility", J.P. Kaminski, D. Enyeart and D. White, Proceedings of the Int. Conf on IR&MM Waves, Naples, Florida (1990).

"Far-infrared cavity dump coupling of the UC Santa Barbara Free-electron Laser", J. Kaminski, J. Spector, C.L. Felix, D.P. Enyeart, D.T. White and G. Ramian, Appl. Phys. Lett. 57, 2770 (1990).

"Graded Potential Wells with Quasi-Uniform Charge Distribution", A. Wixforth, M. Sundaram, D. Donnelly, J.H. English and A.C. Gossard, Surf. Sci., 228, 489 (1990).

"Current Applications Using the UCSB Free-electron Laser", J. Kaminski, J. Nucl. Inst. and Meth. in Phys. Res., A296, N1-3, 784 (1990).

Far Infrared Saturation Spectroscopy of the Mn Impurity Mode in Antiferromagnetic FeF<sub>2</sub>", J. Kaminski and V. Jaccarino, to be published Phys. Rev. B.

"Generation of Subnanosecond High Power Far Infrared Pulses Using a FEL Pumped Passive Resonator", J. Burghoorn, to be published J. Appl. Phys.,

"Far Infrared Perturbation of Electron Tunneling in Reaction Centers?", R.H. Austin and Mi K. Hong, Chem Phys., 158, 473 (1991).

"Photoluminescence from Al<sub>x</sub>Ga<sub>1-x</sub>As/GaAs Quantum Wells Quenched by Intense Far-infrared Radiation", S.M. Quinlan, A. Nikroo, M.S. Sherwin, M. Sundaram and A.C. Gossard, to be published, Phys. Rev.

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